

82247

FORCED OSCILLATIONS OF SUPPORTED DROPS

Edward D. Wilkes and Osman A. Basaran*

School of Chemical Engineering, Purdue University
West Lafayette, IN 47907-1283

ABSTRACT

Oscillations of supported liquid drops are the subject of wide scientific interest, with applications in areas as diverse as liquid-liquid extraction, synthesis of ceramic powders, growing of pure crystals in low gravity, and measurement of dynamic surface tension. In this research, axisymmetric forced oscillations of arbitrary amplitude of viscous liquid drops of fixed volume which are pendant from or sessile on a rod with a fixed or moving contact line and surrounded by an inviscid ambient gas are induced by moving the rod in the vertical direction sinusoidally in time. In this paper, a preliminary report is made on the computational analysis of the oscillations of supported drops that have "clean" interfaces and whose contact lines remain fixed throughout their motions. The relative importance of forcing to damping can be increased by either increasing the amplitude of rod motion A or Reynolds number Re . It is shown that as the ratio of forcing to damping rises, for drops starting from an initial rest state a sharp increase in deformation can occur when they are forced to oscillate in the vicinity of their resonance frequencies, indicating the incipience of hysteresis. However, it is also shown that the existence of a second stable limit cycle and the occurrence of hysteresis can be observed if the drop is subjected to a so-called frequency sweep, where the forcing frequency is first increased and then decreased over a suitable range. Because the change in drop deformation response is abrupt in the vicinity of the forcing frequencies where hysteresis occurs, it should be possible to exploit the phenomenon to accurately measure the viscosity and surface tension of the drop liquid.

1. MOTIVATION, BACKGROUND, AND OBJECTIVES OF RESEARCH

A fundamental understanding of oscillations of both free and supported — pendant or sessile — liquid drops is of practical and scientific importance. A better understanding of oscillations of supported drops, the goal of this research and the subject of this brief research progress report, than that currently available is needed in areas as diverse as: (a) electric field-enhanced liquid-liquid extraction^{1,2}, (b) electrospray methods for the synthesis of simple- and mixed-oxide ceramic precursor powders³, (c) growing of pure crystals in the reduced gravity environment of space⁴, and (d) measurement of dynamic surface tension by the pulsating bubble (drop) technique^{5,6} and the growing drop technique⁷⁻⁹.

In (a) and (b), the energy required to break up and/or atomize supported drops can be minimized by taking advantage of the resonant coupling that can occur between the natural oscillations of the drop and the controlled oscillations of the driver. In (c), it is important to understand the effect of support vibrations on the quality of grown crystals. In a pulsating bubble (drop) surfactometer (PBS) (d), a bubble that is pendant from a tube and is surrounded by a surfactant solution is forced to undergo oscillations to infer the dynamic surface tension of the interface. In the growing drop technique (d), interfacial surface area is created either by impulsively growing out of a capillary tube an initially static pendant drop or repeatedly growing and detaching drops, viz. periodically dripping drops, from a tube by continuously flowing the drop liquid through it. In the experiments of Nagarajan and Wasan⁷, oscillations in drop shape and velocity and pressure fields arise after the static drop is impulsively set in motion. In the continuous flow experiments of MacLeod and Radke⁸ and Zhang *et al.*⁹, similar oscillations occur after one drop detaches from the capillary and another one starts growing from it following the rupture of the liquid bridge connecting the former drop to the rest of the liquid in the capillary (cf. Zhang and Basaran¹⁰). The oscillations of pendant drops that arise during early times in these growing drop experiments not only create an uncertainty in the initial state

*Corresponding author. Phone: 317-494-4061; Fax: 317-494-0805; E-mail: obasaran@ecn.purdue.edu.

of the system but also make it impossible to infer dynamic tension values for times lower than about 20 milliseconds despite the desirability of being able to do so in milliseconds and even in submilliseconds⁹.

However, while the dynamics of oscillating free drops have been studied for over a century since the time of Lord Rayleigh¹¹, oscillations of supported drops had not been studied until recently and have received only limited attention to date. Previous studies of dynamics of supported drops¹²⁻¹⁹ have been highly restrictive, including restriction to small-amplitude oscillations, irrotational oscillations of inviscid drops, and free oscillations. Moreover, in none of these previous studies has the effect of surfactants on finite-amplitude oscillations been considered. Given the widespread occurrence of applications that are enumerated above and the poor state of understanding of them that currently exists, the major goal of this research program is to develop a comprehensive understanding of the finite-amplitude forced oscillations of supported drops that either are pure liquids or contain surface-active species.

For the purpose of illustration, in this brief progress report attention is focused on forced oscillations of drops that are supported on a solid rod, as shown in Fig. 1. It is also taken here that the three-phase contact line where the drop liquid, ambient fluid, and rod meet remains pinned to the sharp edge of the rod throughout the drop motion and the drop is devoid of any surface-active species.

2. MATHEMATICAL FORMULATION

The system is an axisymmetric drop of an incompressible, Newtonian liquid of constant viscosity μ and constant density ρ that is pendant from (or sessile on) a circular cylindrical rod of radius R that lies along the direction of the gravitational acceleration \mathbf{g} , as shown in Fig. 1. The drop is forced to oscillate by moving the rod in the vertical direction sinusoidally in time with frequency $\tilde{\Omega}$ and amplitude \tilde{A} so that the instantaneous position of the rod tip is given by $\tilde{z}' = \tilde{A} \sin \tilde{\Omega} \tilde{t}$ where \tilde{z}' is axial distance measured in an inertial frame of reference in the direction of (opposite to) gravity for a pendant (sessile) drop and \tilde{t} is time. The ambient fluid surrounding the drop has negligible density and viscosity and exerts a constant pressure and negligible drag on the drop as it oscillates. The drop/ambient fluid interface has constant surface tension σ . Throughout the motion, the three-phase contact line (circle) remains pinned to the edge of the face of the circular rod.

The problem is cast onto a moving frame of reference in which the rod is stationary by the transformation $\tilde{z} = \tilde{z}' - \tilde{A} \sin \tilde{\Omega} \tilde{t}$. The equations, boundary conditions, and initial conditions that govern the dynamics are put in dimensionless form by using the radius of the rod R as the length scale and the quantity $\sqrt{\rho R^3 / \sigma}$ as the time scale. In what follows, variables with tildes over them are dimensional whereas the same variables without tildes are dimensionless.

The dimensionless groups that govern the forced oscillations of supported drops are (1) a Reynolds number $Re \equiv (1/\nu)\sqrt{\sigma R/\rho}$, (2) a gravitational Bond number $G \equiv \pm \rho g R^2 / \sigma$, (3) dimensionless forcing amplitude A , (4) dimensionless forcing frequency Ω , and (5) dimensionless drop volume, which is parametrized by a parameter α that varies between -1 and 1 such that $\alpha = 0$ corresponds to a drop whose volume equals that of a hemisphere.

In this paper, two different types of initial conditions are considered. In one of these, a supported drop that is in static equilibrium for times $t \leq 0$ is impulsively set into oscillation by moving the rod in the vertical direction sinusoidally in time for all time $t > 0$ with a fixed forcing frequency Ω and amplitude A . The transient Navier-Stokes system is then integrated in time until the drop motion approaches a time-periodic steady state or a limit cycle. With the other initial condition, once the drop attains a steady oscillatory state at frequency $\Omega = \Omega_1$, the forcing frequency is then changed by a finite amount to $\Omega = \Omega_2 = \Omega_1 + \Delta\Omega$. The transient system is then integrated until a new steady oscillatory state is reached. This procedure is then repeated by incrementing the forcing frequency. Indeed, frequency sweeps are carried out such that the forcing frequency is first increased over a range $\Omega_{low} \leq \Omega \leq \Omega_{high}$ and then decreased over the same range.

3. RESULTS

Figure 2 shows the drop aspect ratio a/b , the ratio of the length of the drop along the axis of symmetry to the rod radius, and the instantaneous location of the rod tip in a fixed frame of reference after the drop has reached a state of steady oscillations during approximately one period for the situation in which the

equilibrium drop shape is a hemisphere ($\alpha = 0$), $Re = 10$, $G = 0$, and $A = 0.1$. The forcing frequency $\Omega = 4$ so that the period of steady oscillations is $\pi/2$. The out of phase oscillations between the rod and the drop aspect ratio give rise to interesting fluid motions inside the drop (not shown but see Wilkes and Basaran²⁰) in particular near maximum and minimum drop deformations and are due to the differences in the time scale of vorticity diffusion from the solid surface and the time scale of rod motion. This out of phase motion between the rod and the liquid underneath the fluid interface has also been observed by Chen and Tsamopoulos²¹ in the forced oscillations of liquid bridges.

Whereas the eigenfrequency of infinitesimal amplitude oscillations is independent of the disturbance amplitude, increasing the forcing amplitude decreases the resonance frequency²⁰. The downward shift of the resonance frequency with increasing forcing amplitude shows that oscillating supported drops exhibit a *soft* nonlinearity.

Were surface tension, density, and rod radius held fixed, a change in Re reflects a change in the viscosity of the drop liquid. Figure 3 shows the effect of Re on the variation of the maximum aspect ratio achieved during steady oscillations, $(a/b)_m$, with forcing frequency when $\alpha = 0$ and $G = 0$ for drops that are forced to oscillate at a value of the forcing amplitude fixed at $A = 0.10$. As with simple systems such as the Duffing oscillator, the results highlighted show that as viscosity (Re) decreases (increases), the drop deformation increases and the resonance frequency decreases. Figure 3 shows that oscillation modes, in particular modes other than the primary oscillation mode, become easier to detect as Re increases. Within the range of forcing frequencies examined, a second peak in $(a/b)_m$ could not be detected for $Re \leq 5$ and no peaks could be detected for $Re = 1$.

During nonlinear oscillations of supported drops, as Re increases the dissipation of energy by viscous forces decreases relative to the the input of kinetic energy into the drop due to the rod motion which manifests itself as an increase in fluid inertia. As in the case of increasing forcing amplitude discussed previously, one can think of this roughly as increasing the relative importance of forcing to damping. The soft nonlinearity of the system is once again made plain by Fig. 3, which shows that as the forcing to damping ratio increases, the observed peaks in deformation amplitude $(a/b)_m$ are skewed to the left or lower values of the forcing frequency. It is well known for the Duffing oscillator that as the ratio of forcing to damping further rises, the ascending side of the Duffing profile can actually turn back on itself. This yields a range of forcing frequencies over which two stable limit cycles exist. This jump phenomenon, also known as hysteresis, is exhibited by many nonlinear systems. For example, DePaoli *et al.*²² have reported experimental observations of hysteresis in drop response for pendant drops that are forced to oscillate by carrying out frequency sweeps in which the forcing frequency is first increased over a range and then decreased over the same range. By continuously varying the forcing frequency, DePaoli *et al.* were able to observe both limit cycles.

Next the response of pendant drops to frequency sweeps were studied. The effect of increasing forcing amplitude on the dynamics of a pendant drop at the highest value of Re shown in Fig. 3 was investigated. When the forcing amplitude was sufficiently small, the computed value of $(a/b)_m$ for increasing values of Ω was the same as that for decreasing values of Ω . However, Fig. 4(a) shows that the drop response is hysteretic when the forcing amplitude is increased to 0.05. Fig. 4(b) shows the phase portraits of the drop in the plane of velocity of the drop tip (v_0) versus position of the drop tip (f_0) at the same value of the forcing frequency albeit the portrait on the left corresponds to the upward part of the sweep whereas the one on the right corresponds to the downward part of the sweep. The phase portraits of Fig. 4(b) plainly demonstrate the existence of two stable limit cycles.

4. CONCLUSIONS AND OUTLOOK

The abruptness of the transition in drop deformation at a well defined value of the forcing frequency that is typical of hysteretic response offers a potentially highly accurate method for making physical property measurements with oscillating supported drops and liquid bridges although no attempt has heretofore been made to take advantage of such drop¹⁹ or bridge response²¹.

Oscillating bubbles and drops are widely used in measuring dynamic surface tension of gas-liquid interfaces (see, e.g., Chang & Franses^{5,6}). However, the underlying flow field is always simplified in these studies to facilitate the solution of the surfactant transport problem. The methods and results reported in this

paper form the necessary preliminaries for analyzing rigorously the combined problems of flow and surfactant transport in and around oscillating drops and bubbles for making improved dynamic surface tension measurements. Such analyses and the requisite accompanying experimental studies are now underway in our laboratory.

REFERENCES

- ¹T. C. Scott and R. M. Wham, "Surface area generation and droplet size control in solvent extraction systems utilizing high intensity electric fields," U. S. Patent 4,767,515 (August 30, 1988).
- ²K. J. Ptasinski and P. J. A. M. Kerkhof, "Electric field driven separations: phenomena and applications," *Sep. Sci. Technol.* **27**, 995 (1992).
- ³M. T. Harris, T. C. Scott, and C. H. Byers, "Method and apparatus for the production of metal oxide powder," U. S. Patent 5,122,360 (June 16, 1992).
- ⁴M. Strani and F. Sabetta, "Free vibrations of a drop in partial contact with a solid support," *J. Fluid Mech.* **141**, 233 (1984).
- ⁵C-H. Chang and E. I. Franses, "An analysis of the factors affecting dynamic tension measurements with the pulsating bubble surfactometer," *J. Colloid Interface Sci.* **164**, 107 (1994).
- ⁶C-H. Chang and E. I. Franses, "Dynamic tension behavior of aqueous octanol solutions under constant-area and pulsating-area conditions," *Chem. Eng. Sci.* **49**, 313 (1994).
- ⁷R. Nagarajan and D. T. Wasan, "Measurement of dynamic interfacial tension by an expanding drop tensiometer," *J. Colloid Interface Sci.* **159**, 164 (1993).
- ⁸C. A. MacLeod and C. J. Radke, "A growing drop technique for measuring dynamic interfacial tension," *J. Colloid Interface Sci.* **160**, 435 (1993).
- ⁹X. Zhang, M. T. Harris, and O. A. Basaran, "Measurement of dynamic surface tension by a growing drop technique," *J. Colloid Interface Sci.* **168**, 47 (1994).
- ¹⁰X. Zhang and O. A. Basaran, "An experimental study of dynamics of drop formation," *Phys. Fluids* **7**, 1184 (1995).
- ¹¹J. W. S. Rayleigh, "On the capillary phenomena of jets," *Proc. R. Soc. Lond., Ser. A*, **29**, 71 (1879).
- ¹²H. Rodot, C. Bisch, and A. Lasek, "Zero gravity simulation of liquids in contact with a solid surface," *Acta Astronaut.* **6**, 1089 (1979).
- ¹³C. Bisch, A. Lasek, and H. Rodot, "Comportement hydrodynamique de volumes liquides spheriques semi-libres en apesanteur simulée," *J. Méc. Théor. Appl.* **1**, 165 (1982).
- ¹⁴C. Bisch, "Les modes de vibrations axiales et tangentielles d'une sphère semi-libre," *C. R. Acad. Sc. Paris* **293**, 107 (1981).
- ¹⁵C. Bisch, "Axial and tangential vibrational modes for semi-free liquid spheres in microgravity nodal circles and nodal points position," *35th Cong. Intl. Astronautica Federation*, Paper No. IAF-84-138, Lausanne, Switzerland, 1984.
- ¹⁶M. Strani and F. Sabetta, "Viscous oscillations of a supported drop in an immiscible fluid," *J. Fluid Mech.* **189**, 397 (1988).
- ¹⁷N. E. Bixler and R. E. Benner, "Finite element analysis of axisymmetric oscillations of sessile liquid drops," *Proc. 4th Intl. Conf. Numer. Meth. Laminar Turbulent Flow* (Swansea, UK, 1985), p. 1336.
- ¹⁸T. Tsukada, M. Sato, N. Imaishi, M. Hozawa, and K. Fujinawa, "A theoretical and experimental study on the oscillation of a hanging drop," *J. Chem. Eng. Japan* **20**, 88 (1987).
- ¹⁹O. A. Basaran and D. W. DePaoli, "Nonlinear oscillations of pendant drops," *Phys. Fluids* **6**, 2923 (1994).
- ²⁰E. D. Wilkes and O. A. Basaran, "Forced oscillations of pendant (sessile) drops," submitted to *Phys. Fluids* (1996).
- ²¹T-Y. Chen and J. A. Tsamopoulos, "Nonlinear dynamics of capillary bridges: theory," *J. Fluid Mech.* **255**, 373 (1993).
- ²²D. W. DePaoli, J. Q. Feng, O. A. Basaran, and T. C. Scott, "Hysteresis in forced oscillations of pendant drops," *Phys. Fluids* **7**, 1181 (1995).

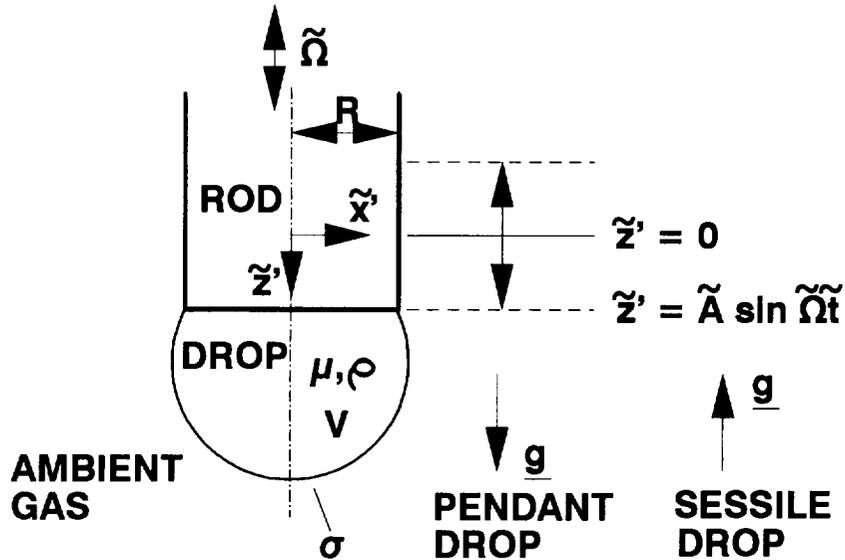


Figure 1. A liquid drop that is pendant from or sessile on a solid rod and undergoing forced oscillations in a vacuum or a gas of negligible density and viscosity.

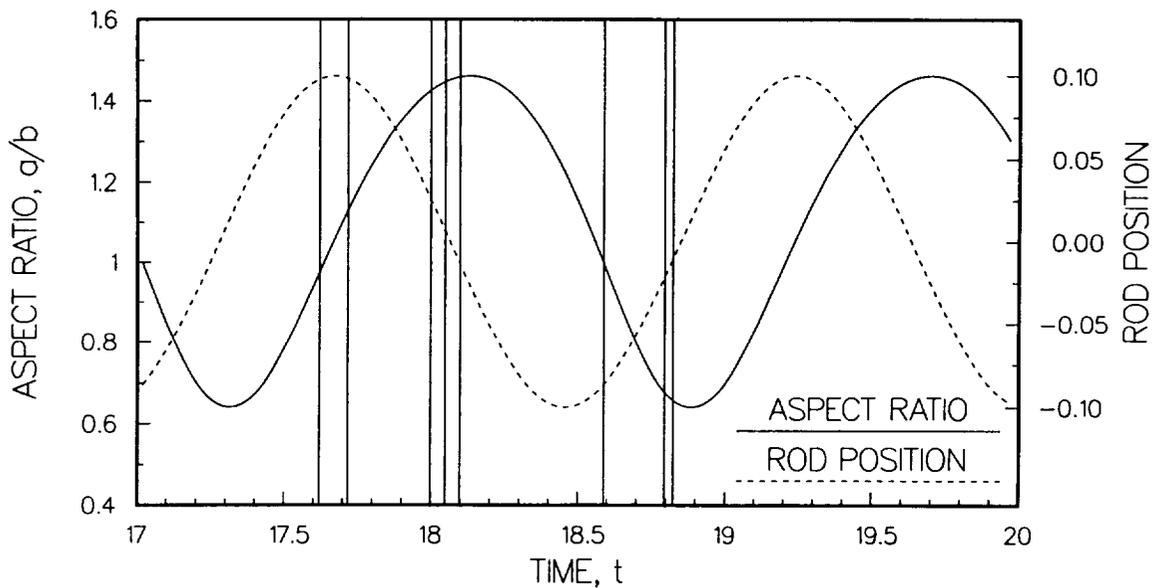


Figure 2. Variation in time of the drop aspect ratio and position of the rod tip. The vertical lines indicate times at which the flow field undergoes interesting transitions (see reference 20). For example, at $t = 17.620$ and 18.098 fluid particles inside the drop are following the motion of the rod. However, due to finite inertia, at $t = 17.717$ and 18.588 , some fluid particles are moving in the direction of the rod whereas others are moving in the opposite direction. At the remaining times, recirculating eddies are visible inside the drop.

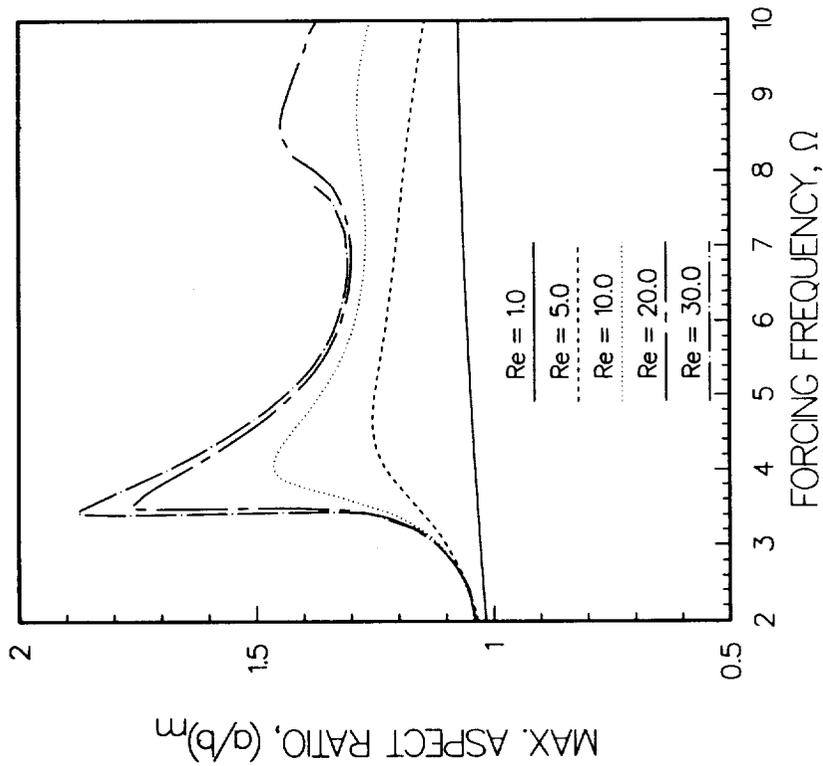
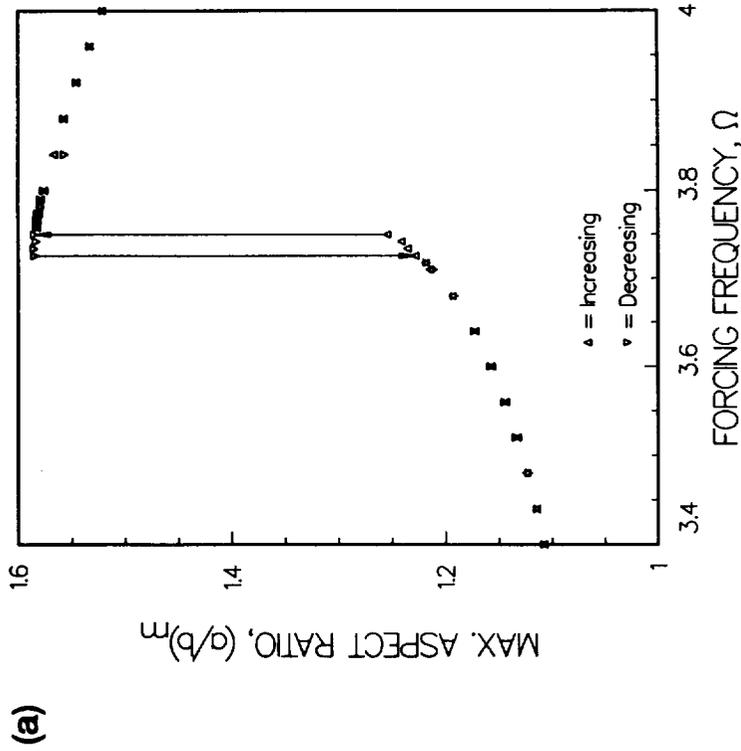
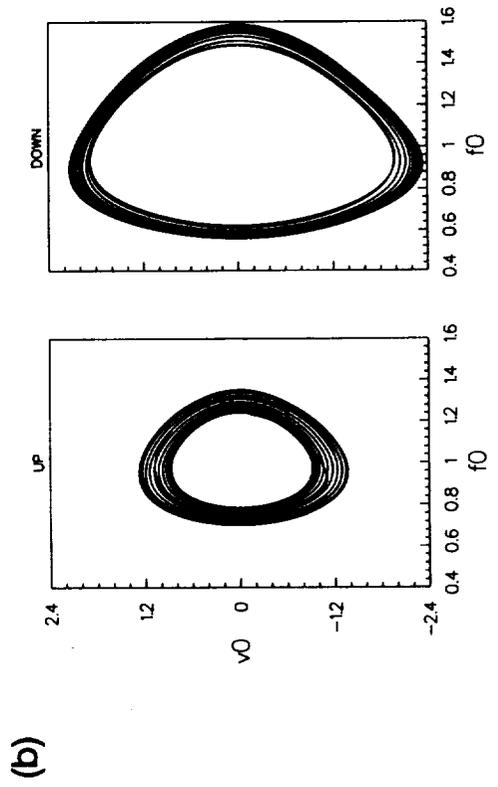


Figure 3. (Top) Effect of Reynolds number on the variation of the aspect ratio at maximum drop deformation during steady-state oscillations with the forcing frequency. The first resonance frequency equals 4.567 when $Re=5$, 4.079 when $Re=10$, 3.534 when $Re=20$, and 3.469 when $Re=30$.



(a)



(b)

Figure 4. (Right) (a) Drop response to a frequency sweep. (b) Phase portraits at $\Omega=3.742$, showing the two limit cycles. In both (a) and (b), $Re=30$, $G=0$, $\alpha=0$, and $A=0.05$.